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Phase-field Simulation Studies of Multiple Grains Coupled With Force Flow Field*

Li Feng[1,2,a*] Lingmin An[1,a] Changsheng Zhu[1,a] Yang Lu [1,a] Rongzhen Xiao [1,2,a]

and Bo Cheng[1,a]

[1]Institute of Material Science and Engineering, Lanzhou University of Technology, Lanzhou, China

[2]Gansu Non-ferrous Metals and Composite Materials Engineering Technology Research Center, Lanzhou China

[a]fenglils@lut.cn

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Abstract. Coupling the force flow field with the phase field model for the isothermal growth of dendrite multiple grains, Sola algorithm is used to calculate the flow speed and pressure of liquid metal, double grid numerical method was used to reduce the calculation amount of computer simulation, the space factor and time factor were introduced to improve the accuracy of double grid numerical calculation, Taking Al-2%-Cu alloy for example, the dendrite growth process of the binary alloy was simulated under forced convection environment; Simulation results can capture the real dendrite growth and interactions of the liquid metal flow in the process of dendrite growth under forced convection environment: The flow of metal liquid affects the growth morphology of dendrite multi-grain. The flow of liquid metal changes the growth speed of dendrite tip in different directions for each dendrite, the greater of the liquid metal initial flow speed, the worse of dendrite morphology symmetry; different initial flow speeds result in the different distance between each dendrite. The metal liquid forced flow causes the instability of dendrite growth interface, it changes the degree of undercooling, composition, dendrite tip growth rate and the curvature radius of dendrite tip in the dendrite growth forefront interface. For the forefront dendrite growth interface free energy system, the dendrite tip growth speed and curvature radius were adjusted by the competitive growth of dendrite tips, thus reached a new stable state for the interface, which resulted in the emergence of bifurcation in dendrite tips. The liquid metal flow speed between different grains was affected by the relative position and morphology between different grains, and also affected by the initial inflow speed of the liquid metal.

Introduction

As a typical non-equilibrium self-organization phenomenon, dendrite growth has been one of the important research topics in condensed matter physics [1]. Phase-field method is a novel microstructure simulation method, which not only avoids the difficulty of tracking complex interface, but also provides an open theoretical framework for describing the evolution of the microstructure [2,3]. Coupling phase field with temperature field, solute field, flow field and other external field, the dendrite evolution process during solidification can be directly simulated, and the effects of solid-liquid interface curvature effects, kinetic effects, interference, aeolotropy as well as other external fields on the solidification microstructure formation can be systematic studied [4].

At present, there are many researchers at home and abroad have simulated the microstructure during solidification using phase-field method, the phase-field method simulation coupling flow field during the solidification process also made some achievement.

Tong and Beckermann [5] proposed the phase-field model considering the flow field and conducted quantitative simulation, Lan and Shin [6] simulated the effects of forced convection on Ni-Cu binary alloy non-isothermal free dendrite, Nestler et al[7] studied the effects of convection on monotectic eutectic lamellar space using the phase-field simulation, Li and Beckermann [8] established a static film model, the effect of flow on the alloy dendrite growth, and the model was compared with the available experimental data of succinonitrile-acetone alloy. Rezende et al[9] simulated the dendrite morphology and solute segregation of Fe-Mn-C austenitic steels during solidification in flowing environment by phase-field simulation, the simulation results and the corresponding experimental results have confirmed that, in the alloy, Mn showed serious segregation tendency. LONG Wen-yuan et al [10] simulated the effects of forced convection on the free non-isothermal dendrite of Al-Cu binary alloy.

Because the large amount of calculation is existed in phase-field simulation coupling flow field, calculation method is one of the hot spots in the field. Jeong et al [11] used a parallel algorithm, simulated the effects of flow on the three-dimensional dendrite growth by using adaptive finite element method, Tönhardt and Amberg [12] simulated the dendrite growth with adaptive finite element method under natural convection and forced convection, Lu et al [13] employed multi-grid method, solving phase-field equations and the energy equation using the finite difference method, quantitatively simulated the effect of force convection on the three-dimensional dendrite growth, Dong et al [14] proposed an efficient and effective time-step method for studying two-phase flows, it completely de-couples the computations of the velocity, pressure involving large density ratios and interfacial topology changes.

In the metal solidification process, the flow field which influences the evolution of dendrite will change with the impact of dendrite evolution, therefore, in order to simulate the evolution of dendrite accurately during solidification, coupling the changing flow field is necessary. Due to the limitation and impact of large calculation amount and other factors, the above part of the study only explored the impact of flow field on dendrite growth, but the impact of dendrite on flow field is less mentioned. In this paper, the isothermal solidification dendrite growth model coupling the forced flow-field for binary alloy was established, calculating the phase field and solute field equations by the finite difference method; the flow field equations were calculated by Sola algorithm; in order to save calculation time, reduce the computational volume, double grid method was adapted to calculate the phase field, solute field equations and flow field equations, respectively; take Al-Cu binary alloy for example, the simulation results can capture the interactions of the liquid metal flow and multiple grains dendrite growth under forced convection environment.

Phase-field model for dendrite growth

Phase field equation and solute field equation

In this article, the binary alloy multi-grains dendrite growth isothermal phase-field model which Feng Li et al [15] proposed was coupled with the flow field, the phase-field equation can be expressed as equation (1):

$$\frac{\partial \phi}{\partial t} = M\left[\varepsilon^2(\theta_i)\nabla^2\phi - f_\phi\right] \tag{1}$$

Where, M is solid interface migration rate, Φ is phase field order parameter, t is time variable, f_Φ represents the first derivative of free energy density on the phase-field order parameter, and its representation form is as equation (2):

$$f_\phi = \frac{RT}{V_m} h'(\phi) \ln \frac{(1-c_S^e)(1-c_L)}{(1-c_L^e)(1-c_S)} + Wg'(\phi) \tag{2}$$

Where, R is gas constant, T is temperature, V_m is molar volume, $h(\Phi)$ is potential function, W is phase field parameter, $g(\Phi)$ is residual free energy function, c represents the concentration of solute in the alloy, the subscript L, S represents for liquid and solid phase respectively, the superscript e represents equilibrium state. $\varepsilon(\theta_i)$ is a parameter associated with the interfacial energy, which is expressed in the form of equation (3):

$$\varepsilon(\theta_i) = \varepsilon_0 (1 + \upsilon \cos(k\theta_i)) \tag{3}$$

Where k is anisotropy modulus, v is the anisotropy coefficient, θ_i is the included angle between the interface and the preferential growth direction of a certain crystal [15].

After coupling forced flow field, the solute distribution during solidification process is combined influenced by the solute redistribution, solute diffusion and liquid flow speed, then the solute field equation can be expressed as equation (4):

$$\frac{\partial c}{\partial t} = \nabla\left(\frac{D(\phi)}{f_{cc}} \nabla f_c\right) + \nabla(V \cdot c) \tag{4}$$

Where, $D(\Phi)$ is solute diffusivity, f_c, f_{cc} is the first-order and second-order derivative of free energy to concentrations, $\boldsymbol{V}$ is the velocity vector of liquid flow.

Flow field equations

In the model, the flow field equation consists of mass conservation equation and momentum conservation equation. For the incompressible fluid, the mass conservation equation can be expressed as equation (5):

$$\mathrm{div}(V)=0 \tag{5}$$

Momentum conservation equation can be expressed as equation (6),

$$\begin{cases} \dfrac{\partial V_x}{\partial t} + \mathrm{div}(V_x V) = \mathrm{div}(\nu\, \mathrm{grand}\, V_x) - \dfrac{1}{\rho}\dfrac{\partial p}{\partial x} \\ \dfrac{\partial V_y}{\partial t} + \mathrm{div}(V_y V) = \mathrm{div}(\nu\, \mathrm{grand}\, V_y) - \dfrac{1}{\rho}\dfrac{\partial p}{\partial y} \end{cases} \tag{6}$$

Where, v is the kinematic viscosity of liquid metal, ρ is the density of the liquid metal, p represents pressure, and the subscripts x, y represent the component of variable on x, y axis.

Simulation parameters values and model numerical calculation

The determination of phase-field parameters

Migration rate of solid interface (M) is expressed as equation (7),

$$M^{-1} = \frac{\varepsilon_0^3}{\sigma\sqrt{2W}} \left(\frac{1}{D} \zeta \left(c_L^e, c_S^e\right) \right) \tag{7}$$

Where, ε_0 and W are phase-field parameters, which are calculated by the interfacial energy σ and the thickness of interface λ, they are expressed as equation (8)-(10),

$$\varepsilon_0 = \sqrt{\frac{6\lambda\sigma}{2.2}} \tag{8}$$

$$W = \frac{6.6\sigma}{\lambda} \qquad (9)$$

$$\zeta = \frac{RT}{V_m}\left(c_l^e - c_s^e\right)^2 \times \int_0^1 \frac{h(\phi)\left[1-h(\phi)\right]}{\left[1-h(\phi)\right]c_l^e\left(1-c_l^e\right)+h(\phi)c_s^e\left(1-c_s^e\right)} \bullet \frac{d\phi}{\phi(1-\phi)} \qquad (10)$$

Material parameters

In this paper, take Al-2%mol-Cu alloy for example, the coupled flow field dendrite growth is simulated. Physical parameters of the alloy are shown in Table 1.

Table 1. Material parameters

Material Parameters	Al-2%mole-Cu
Interfacial energy $\sigma/(J\cdot m^{-1})$	0.093
Melting temperature T_m/K	933.3
Latent heat $L/(kJ\cdot kg^{-1})$	389.0
Thermal conductivity $k/(W\cdot(m\cdot k)^{-1})$	192.6
The equilibrium constant k^e	0.14
Liquid solute diffusion coefficient $D_l/(m^2\cdot s^{-1})$	3.0×10^{-9}
Solid solute diffusion coefficient $D_s/(m^2\cdot s^{-1})$	3.0×10^{-13}
Liquidus slope m^e	620
Kinematic viscosity $v/(m^2\cdot s^{-1})$	1.0×10^{-6}

The initial and boundary conditions

In the solidification process, the formation of nucleus is divided into spontaneously homogeneous nucleation and non-spontaneously. In this model, several nucleus of heterogeneous nucleation are placed in the computational domain randomly, assuming the radius is R, then

$$\text{When } x^2 + y^2 \le R^2, \quad \phi = 1$$
$$T = T_m - \Delta T \qquad (11)$$
$$\text{When } x^2 + y^2 > R^2, \quad \phi = 0$$
$$T = T_m - \Delta T \qquad (12)$$

Where, x, y is the horizontal and vertical coordinate of the nucleus center, respectively. T is the dimensional temperature, ΔT is the degree of undercooling, T_m is the melting point. The number of grains takes any number smaller than the maximum nucleation number, the maximum nucleation number can be calculated using the formula for nucleation density [12], and the expression is as equation (13),

$$\frac{dn}{d(\Delta T)} = \frac{n_{\max}}{\sqrt{2\pi}\Delta T_\sigma}\exp\left(-\frac{(\Delta T - \Delta T_{\max})^2}{2\Delta T_\sigma^2}\right) \qquad (13)$$

Where ΔTmax is maximum nucleation undercooling, ΔTσ is standard deviation undercooling, nmax is the maximum nucleation density.

On the left margin of the simulation area, given a inlet flow rate V(Vx， Vy), the

$$\begin{cases} x \le 1, V_x = V_0, V_y = 0, p = 0 \\ x > 1, V_x = 0, V_y = 0, p = 0 \end{cases} \tag{14}$$

In the calculation area, the calculation of phase field Φ and solute field c both adopt adiabatic boundary condition, namely, Zero-Neumann boundary condition. When calculating the flow speed of liquid metal, the solid-liquid interface area in calculation domain is calculated using a moving boundary condition and the upper and lower boundaries of the calculation domain also use the moving boundary condition, the right boundary is taken as the outlet of liquid metal flow.

Numerical Calculation

Correspond to the x and y axis (the horizontal direction is x) in coordinate system, the computational grid number of phase field and solute field both are 1200×1200, the grid size is 1 × 10-8m (Δx = 1 × 10-8m), set the initial nucleus as the ball with the grids number radius R=10. Solve Equation (1) and Equation (3) using explicit finite difference in the same time, with the same time step Δt, Δt and space step Δx (space step Δx=Δy) must meet the following stable condition (15),

$$\Delta t < \min\left\{\Delta x^2 / (4D_L), \Delta y^2 / (4D_L)\right\} \tag{15}$$

Sola algorithm is used for solve equation (5), when calculating the flow field, the time step Δtf and space step Δxf (space step Δxf =Δyf) must meet the following condition (16),

$$\Delta t_f < \min\left\{\Delta x_f \big/ |V_x|, \Delta y_f \big/ |V_y|\right\} \tag{16}$$

Dual grid Method

Sola algorithm use adaptive iterative method calculate the pressure, the initial value of the pressure on each grid node in the calculation area is randomly assumed, based on the initial condition or the previous value, calculating the next time guess value, in order to satisfy the continuity equation, the pressure in each grid must be corrected, the velocity changes caused by the correction are added to the velocity field in the first step, calculate iteratively until the result meet the accuracy requirement. Obviously, if the pressure changes a lot in a time step, the convergence rate must be very slow. For reducing the calculation amount and saving computation time, dual grid method is designed, two different thickness grids are divided in the same calculation domain, i.e., using different space step ($\Delta x \neq \Delta x_f$) to calculate the phase field, solute field and flow field, the flow field is calculated with the coarse grid. Because of different thickness of the grids, resulting in different space and time accuracy of flow field, phase field and solute field numerical solutions within the calculation region, for this reason, space factor N_w and time factor N_t were introduced,

$$\Delta x_f = N_w \Delta x \quad , \quad \Delta t_f = N_t \Delta t \tag{16}$$

$$V_f(I, J) = V(i \cdot N_w, j \cdot N_w) \tag{17}$$

$$V(i \cdot N_w + n, j \cdot N_w) = V(i \cdot N_w, j \cdot N_w) + n[V_f(I+1, J) - V_f(I, J)] \big/ N_w \tag{18}$$

$$V(i \cdot N_w, j \cdot N_w + n) = V(i \cdot N_w, j \cdot N_w) + n[V_f(I, J+1) - V_f(I, J)] \big/ N_w \tag{19}$$

$$V_f(T) = V(t \cdot N_t) \tag{20}$$

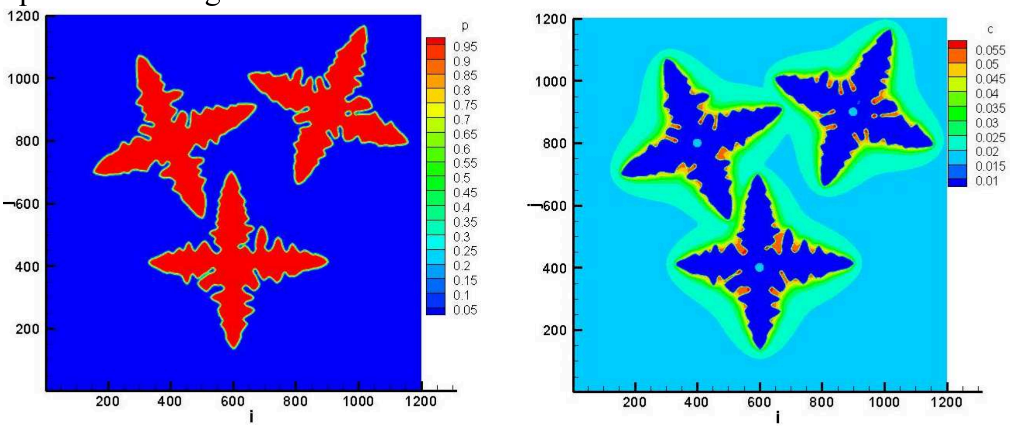
$$V(t \cdot N_t + n) = V(t \cdot N_t) + n[V_f(T+1) - V_f(T)] \big/ N_t \tag{21}$$

Where, V represents the flow velocity, the subscript f indicates the numerical solution of the calculated flow field grid, I, J, T and i, j, t represents the space and time node in different grids, respectively, in formula (17), (18), n is a natural number less than N_w, in formula (20), n is a natural number less than N_t.

Results and Analysis

Effect of external forced flow field on dendrite growth morphology

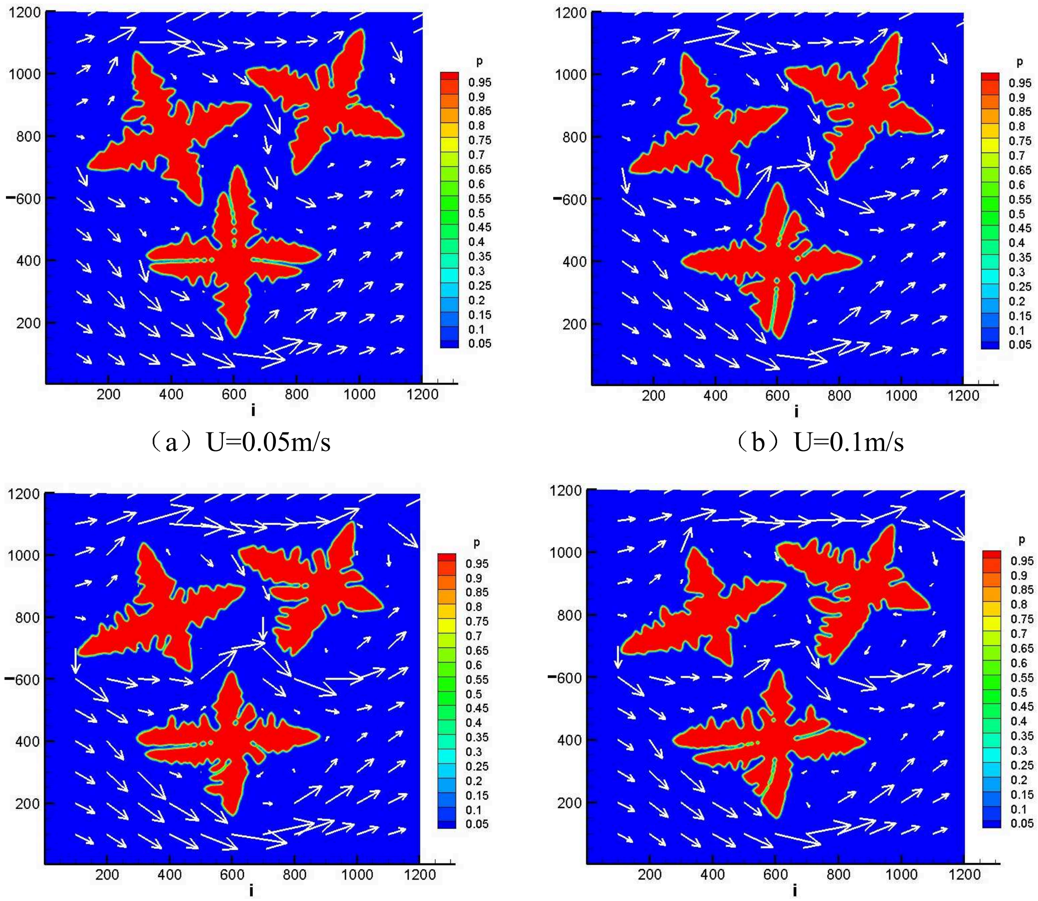
Fig.1 is the simulation results of supercooled liquid metal at 6000Δt, the initial temperature is 900K and in the absence of external forced flow environment: Fig.1 (a) is the dendrite growth morphology of multiple different orientations, Fig.1 (b) is the simulation result of solute distribution within multiple dendrite growth region. In Fig.1 (a), the red area represents for solid phase, blue for liquid, color scale indicates the percentage of solid phase in different colors areas, there is no contact between the different grains at this time and do not produce the mutual influence during the growth process; but when the dendrite grow to contact with each other, their growth will influence each other [12]. The color scale in Fig.1 (b) indicates the solute concentration of different color areas, red represents the high solute concentration and blue indicates low solute concentration.



(a) the morphology of dendrite (b) the distribution of solute

Fig.1 The simulation result of multiple dendrite growth in the absence of external forced flow

Fig.2 is the dendrite morphology simulation results of supercooled liquid metal at 6000Δt, the initial temperature is 900K and in the external forced flow environment: Fig. (a) is the simulation result with an initial flow speed of U=0.05m/s (horizontal direction velocity component is 0.05m/s, the vertical velocity component is 0); in Fig. (b), (c), (d), the initial flow speed is U=0.1m/s, U=0.15m/s and U=0.2m/s, respectively. The arrow direction represents the flow direction of the cold liquid metal and the length of the arrow indicates the velocity of supercooled liquid metal. In the calculation area, the speed of each grid node has been calculated, if the velocity of each node are represented by the arrows, the arrow will be too crowd and make it impossible to show clearly in the picture, in Fig. 2, in order to represent the flow of the undercooling liquid metal better, the density of the arrow is adjusted accordingly.



（a）U=0.05m/s （b）U=0.1m/s

（c）U=0.15m/s （d）U=0.2m/s

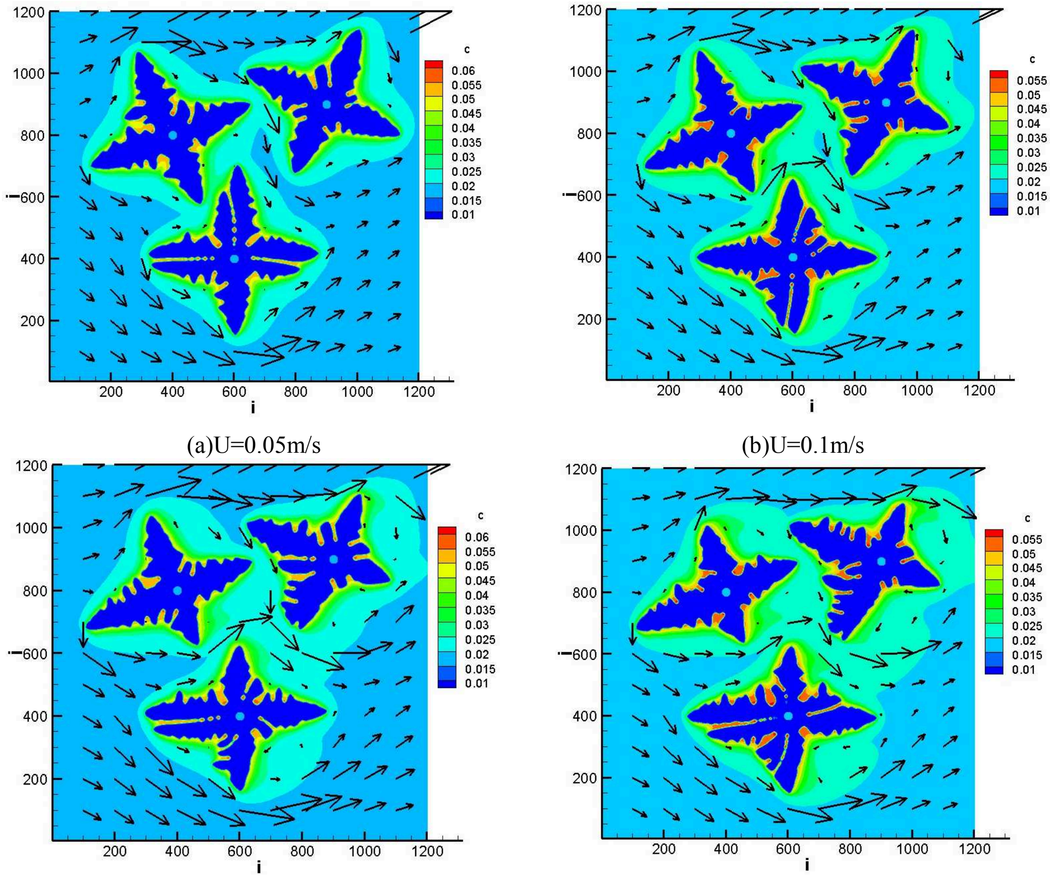
Fig.2 The simulation result of multiple dendrite growth with external forced flow

By contrasting Fig.1 and Fig. 2, it can be confirmed that the initial flow of cold liquid metal will change the morphology of dendrite growth. It can be found from Fig. 2, along with the change of supercooled liquid metal initial flow speed, the dendrite growth morphology changes accordingly, the greater the initial flow speed, the worse symmetry of dendrite morphology; different initial flow speeds result in different space between the dendrites. These phenomena are due to the flow of liquid metal changes the growth speed of dendrite tip in different directions for each dendrite. The impact of liquid metal flow speed on dendrite tip growth rate is more obvious in the single dendrite growth model under the flow environment, and it has been reported [4], so in this paper, it will no longer be discussed.

The flow of liquid metal made the main branches in the below simulation area bifurcating. It can be found from Fig. 2 (a), (b), (c), (d), the effect of liquid metal initial velocity on dendrite tip growth is not monotonic, the greater the initial velocity, the more dendrite tip bifurcation rule is not exist. The free energy on dendrite growth interface front is stable in the pure diffusion condition and it related to the degree of supercooling, composition, dendrite tip growth rate, the dendrite tip curvature radius; the forced flow of liquid metal caused the instability of interface, and changed the degree of supercooling, composition on dendrite growth interface frontier, dendrite tip growth rate and the dendrite tip curvature radius; The free energy system on dendrite growth interface frontier adjust the growth velocity and curvature radius on the dendrite tips through competitive growth of dendrite tips, to reach a new steady state, and therefore dendrite tip bifurcation appeared. This adjustment is achieved by the vibration of free energy in the vicinity of equilibrium positions.

The solute distribution of dendrite under the action of forced flow

Corresponds to Fig.2, Fig.3 is the dendrite growth solute distribution in forced flow environment. In the Figure, different color regions represent the solute concentration in this region, the color scale marks the amount of solute concentration by different colors. The arrow direction represents the flow direction of the cold liquid metal and the length of the arrow indicates the velocity of supercooled liquid metal. Comparing the solute distribution in Figures 3 and 1 (b), it can be found that in simulation area, the solute distribution concentration between the grains on the right and below increases as the increase of flow velocity. This is because the flow of liquid metal brings the solute on dendrite upstream region to this region, these solutes are precipitated from the upstream grains under the action of solute redistribution during the solidification process. In Fig.1 (b), after the precipitation of solute from the grains, there is no liquid flow to take solute away, the solute diffuse freely in the grain growth interface forefront and a thickness diffusion layer formed. In Fig.3, the morphology and thickness of solute in the diffusion layer of the grain growth interface is changed by the liquid metal flow. From Fig.3 (a), (b), (c), (d), the greater the initial velocity of the liquid metal, the larger change of the morphology and thickness of solute in the diffusion layer of the grain growth interface.



(a)U=0.05m/s (b)U=0.1m/s

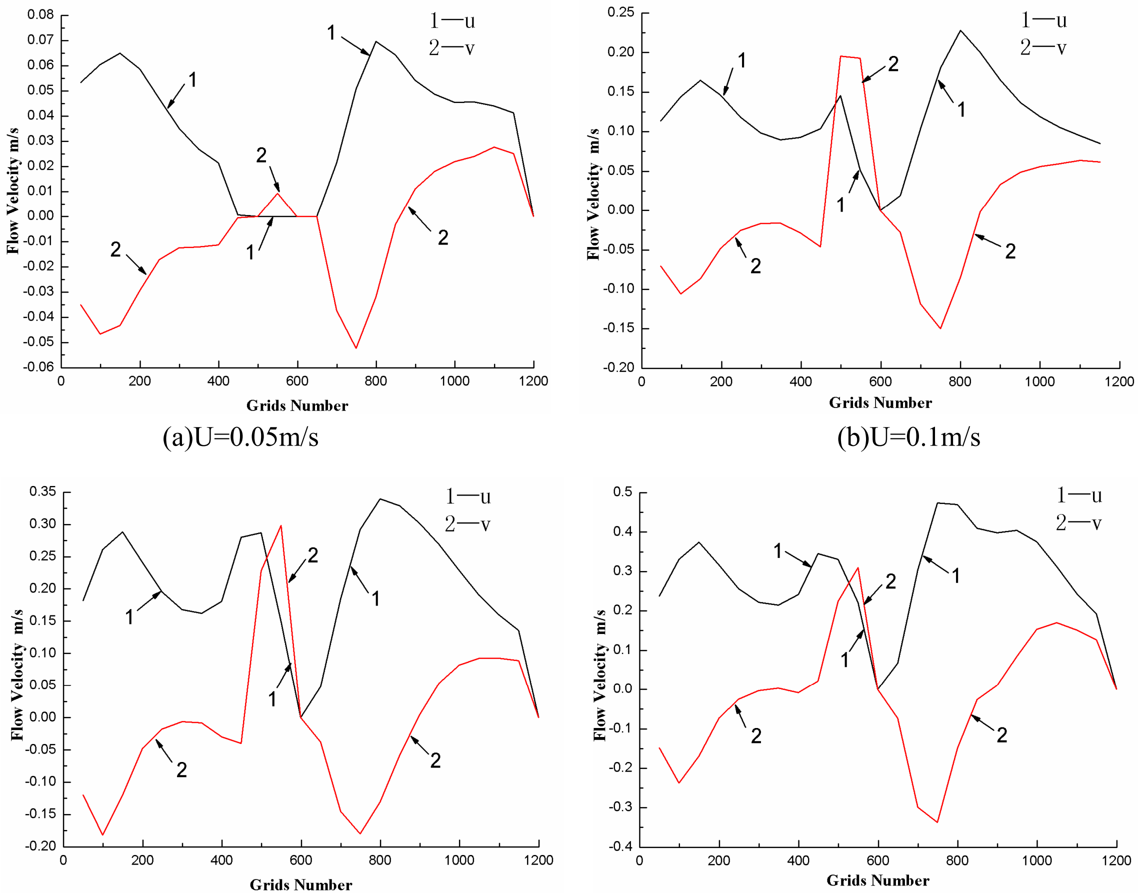
(c)U=0.15m/s (d)U=0.2m/s

Fig.3 The simulation result of multiple dendrite solute distribution with external forced flow

Flow velocity changes of the liquid metal

Dendrite growth initial condition is given a liquid metal inlet velocity $V(Vx, Vy)$ on the left margin of the simulation area, for details see the content of 2.3. Over time, the liquid metal flow speed changes accordingly, and the arrows in Fig.3 and Fig.2 represent the flow direction and size of liquid metal.

Fig.4 corresponds to Fig.2, in Fig. 2, for the liquid metal velocity curves in the region whose ordinate is 600, *u* represents *Vx* (the component of liquid metal flow rate in *X* direction), *v* represents *Vy*(the component of liquid metal flow rate in *Y* direction).



(a)U=0.05m/s (b)U=0.1m/s

(c)U=0.15m/s (d)U=0.2m/s

Fig. 4 Liquid metal flow rate curve

The ordinate indicates the size of the liquid metal flow rate, the negative sign indicates the flow direction is in the opposite. The direction of *u* to the right is set positive, and to left is negative; *v* is positive upward and the negative downward. The abscissas in Fig.4 (a), (b), (c), (d) correspond to that in Fig.2. In Fig. 4 (a), (b), (c), (d), there is region u and v are 0, corresponding to the region in Fig. 2, it can be seen that it is a solid phase region, no liquid, the flow speed is zero.

In Fig.4, the large velocity absolute value area is close to the solid crystal region, and the corresponding abscissa grid number is between 700 and 800. Corresponding to Fig.2, it can be found that the area corresponds to the liquid metal region between upper right dendrite and below dendrite, the arrows on the region is the longest in Fig.2. When the horizontal grid number is above 1100, the absolute value of velocity is gradually reduced to a value close to the initial value. This is caused by the momentum and mass conservation of liquid metal flow, the flow channel cross-sectional area between different grains is small, so the liquid metal flow fast; in the downstream dendrite growth area, the cross-sectional area of liquid metal the passage become larger, result in the reduce of flow velocity. Of course, the relative position between the different grains has a great influence on flow velocity; e.g., the flow speed between upper-left and upper-right dendrites is different from that of between upper-right and below dendrites, this can be manifested by the velocity and direction arrows in Fig.2. When the initial flow speed is low, the flow speed between different grains is smaller, e.g. Fig 4 (a), the absolute value of the velocity component is about 0.07m/s; when the initial flow speed is relatively large, the flow speed between different grains is large, and e.g. Fig.4 (d), the absolute value of the velocity component is about 0.4m /s. This indicates that, the liquid metal flow speed between different grains affected by the relative position and morphology between different grains, furthermore, it also affected by the initial inflow velocity.

Conclusions

(1) Coupling the force flow field with the phase field model for the isothermal growth of dendrite multiple grains, Sola algorithm is used to calculate the flow speed and pressure of liquid metal, double grid numerical method was used to reduce the calculation amount of computer simulation, the space factor and time factor were introduced to improve the accuracy of double grid numerical calculation, Taking Al-2%-Cu alloy for example, the dendrite growth process of the binary alloy was simulated under forced convection environment;

(2) The flow of metal liquid affects the growth morphology of dendrite multi-grains. The greater of the liquid metal initial flow speed, the worse of dendrite morphology symmetry; different initial flow speeds result in different distance between each dendrite. This is because the flow of liquid metal changes the growth speed of dendrite tip in different directions for each dendrite.

(3) The flow of liquid metal made the main branches in the below simulation area bifurcating. The effect of liquid metal initial velocity on dendrite tip growth is not monotonic, the greater the initial velocity, the more dendrite tip bifurcation rule is not exist. The forced flow of liquid metal caused the instability of interface, and changed the degree of supercooling, composition, dendrite tip growth speed and the dendrite tip curvature radius on dendrite growth interface frontier; the free energy system on dendrite growth interface frontier adjust the growth velocity and curvature radius on the dendrite tips through competitive growth of dendrite tips, to reach a new steady state, and therefore dendrite tip bifurcation appeared. This adjustment is achieved by the vibration of free energy in the vicinity of equilibrium positions.

(4) The solute diffusion of the dendrite growth interface is changed by the liquid metal flow, the greater the initial velocity of the liquid metal, the larger change of the morphology and thickness of solute in the diffusion layer of the grain growth interface.

(5) The liquid metal flow speed between different grains affected by the relative position and morphology between different grains, furthermore, it also affected by the initial inflow velocity.

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